CORE
Metadata, citation and similar papers at core.ac.uk
Provided by Elsevier - Publisher Connector

Chinese Journal of Aeronautics 24 (2011) 823-828

Contents lists available at ScienceDirect

Chinese Journal of Aeronautics

journal homepage: www.elsevier.com/locate/cja

# Microstructure and Magnetic Anisotropy of FeCoNbB Films

YANG Youran, WANG Yanyan, LI Ying, BI Xiaofang*

*Key Laboratory of Aerospace Materials and Performance (Ministry of Education), School of Materials Science and Engineering, Beihang University, Beijing 100191, China*

Received 29 November 2010; revised 9 December 2010; accepted 4 January 2011

**Abstract**

Effect of Nb contents on microstructure and magnetic properties of FeCoNbB films has been studied. X-ray diffraction (XRD) analysis reveals that Nb plays a role in refining grain and in facilitating formation of amorphous structure. When Nb content is more than 8 at%, the films transform from crystalline to amorphous structure, accompanied by variations in magnetic properties. An unusual out-of-plane anisotropy component is consequently observed. It can be suggested that the anisotropy induced by increasing Nb contents be attributed to stress-related magnetoelastic anisotropy. The undesirable anisotropy is greatly reduced by thermal annealing, reducing film thicknesses or applying an external magnetic field together with Si addition.

*Keywords:* FeCoNbB films; magnetic anisotropy; Nb contents; annealing; film thickness

## 1. Introduction

Ferromagnetic films have attracted considerable interest for promising applications in micro-devices which can satisfy the requirements of miniaturization and integration in avionics devices such as sensors, transformers, thin film inductors, magnetic flux amplifier and recording system [1-2]. To ensure high-quality performance, soft magnetic materials are required to possess high saturation flux density together with low coercivity [3]. As compared to traditional Fe-based and Co-based materials, FeCo-based alloys are much more favored due to their much higher saturation magnetization, low coercivity and large uniaxial anisotropy [4-5]. Many studies have been conducted on FeCoB alloys, one of the most promising alloys for the applications of recording systems in many fields such as aircraft industries [6-7]. In addition, previous researches have shown that Nb is inclined to be enriched around phase boundaries, and has an effect on decreasing grain size in crystalline alloys [8-10]. On the other hand, magnetic properties are generally dependent on structure transition which is inclined to give rise to anisotropy in soft magnetic materials. However, the presence of out-of-plane spin alignment is undesirable for those to be applied in micro-devices due to introduction of noise [7].

Many reports have suggested that the out-of-plane anisotropy results from reduced symmetry at substrate/film interface, internal stress or columnar growth [1,11-12]. Yu [7] and Coisson [13] found that perpendicular anisotropy disappeared because of annealing and in turn soft magnetic properties were exhibited. Internal stress was suggested to play an important role in the spin reorientation in films. On the other hand, it was observed that columnar structure or texture has a significant influence on spin orientation in some other films, such as NiW [14], FeZrN [15] and FeB [16]. Theoretical calculation made by Chezan [15] also showed that magnitude of magnetoelastic coupling effect was much smaller as compared to perpendicular anisotropy constant in the films.

In this paper, microstructure and anisotropy for Fe-

*Corresponding author. Tel.: +86-10-82315999.
*E-mail address:* bixf@buaa.edu.cn
Foundation item: National Natural Science Foundation of China (50941046)

1000-9361 © 2011 Elsevier Ltd. Open access under CC BY-NC-ND license.
doi: 10.1016/S1000-9361(11)60097-6

CoNbB with various Nb contents is studied for the purpose of obtaining a better understanding of underlying origin of the anisotropy. Furthermore, the dependence of the anisotropy on annealing, thickness and Si addition is also presented in order to find an effective way of eliminating the undesired anisotropy for magnetic softness.

## 2. Experimental

FeCoNbB films were prepared by DC-magnetron sputtering on Si (100) substrates at room temperature. For the deposition, $Fe_{47.5}Co_{47.5}B_5$ alloy was used as target, on which Nb and Si chips were placed. Nb and Si contents were varied by changing the number of chips. Deposition was performed by using high purity argon as sputtering gas. During deposition, the argon pressure was in the range of 0.8-1.3 Pa and sputtering power was fixed at 28.8 W. During field-deposition, an external magnetic field of 40 Oe (1 Oe=80 A/m) was applied along film surface. Films with thicknesses of 140, 210, 420 nm were prepared. As-deposited films were annealed in vacuum for 60 min at the temperature of 100, 150, 200 and 250 °C, respectively. Structure and composition of the films were investigated by X-ray diffraction (XRD, Rigaku, D/MAX2200PC) analysis and energy dispersive spectroscopy, respectively. Vibrating sample magnetometer (Riken Denshi, BHV-55) was used to measure the magnetic properties, by applying magnetic fields parallel to the film plane. Cosine curves of $M$-$\theta$ ($M$ is magnetization, $\theta$ the angle between a certain line on the sample and external magnetic field) were measured for square-shaped samples to determine easy and hard axes. Magnetization represented in this work has been normalized with respect to saturation magnetization.

## 3. Results and Discussion

### *3.1. Effect of Nb contents on microstructure of FeCoNbB films*

Fig. 1 shows XRD patterns of 420 nm-thick FeCoNbB films with different Nb contents. Crystalline diffraction peaks are clearly seen for the films containing 5 at%-7 at% Nb, indicating that the as-deposited films are in a crystalline state. The average grain size for the films can be estimated by Scherrer formula using full width at half maximum (FWHM) value obtained from XRD diffraction peaks, as shown in the inset of Fig. 1. The average grain size shows a slight decrease from 23.5 nm to 22.5 nm with increasing Nb content from 5 at% to 7 at%. The result indicates that grain growth of crystalline phase can be more or less suppressed by increasing Nb contents, as observed in other alloy systems [8,17].

Note that diffraction angle of FeCo(110) peak shifts towards lower angles as compared to that of bulk counterpart as Nb content reaches 7 at%, implying that

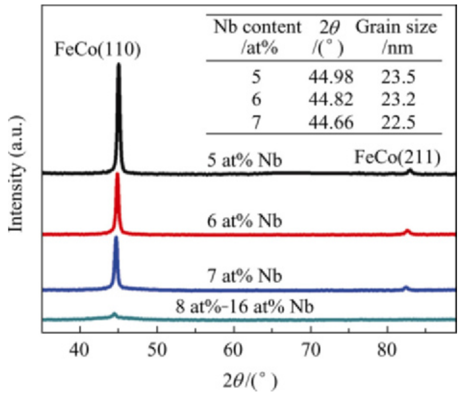


Fig. 1 XRD patterns for films with different Nb contents. Inserted are variations of diffraction angle and grain size with Nb contents.

lattice spacing of the (110) plane was expanded. The expansion of lattice due to Nb addition suggests that the film is inclined to transit from tensile to compressive state. As Nb contents increase to no less than 8 at%, as shown in Fig. 1, a typical broad peak is observed for the films. It indicates that films are in an amorphous state, which can be understood by the role of Nb in suppressing growth of crystalline phase and in turn enhancing glass-forming ability [9].

### *3.2. Effect of Nb contents on magnetic properties of FeCoNbB films*

Variations of magnetic properties with the structure transition have been studied. Fig. 2 presents in-plane hysteresis loops for the films of thickness of 420 nm with Nb contents ranging from 5 at% to 16 at%, where $M_s$ is the saturation magnetization. It is seen that hysteresis for the alloys with Nb contents less than 6 at% is characteristic of soft magnetic properties, showing coercvity lower than 6.5 Oe. By contrast, as Nb content is increased to 8 at%, a great variation in shape of the hysteresis loop is observed. Coercivity of the alloys is increased simultaneously. It is also seen that the hysteresis loop consists of two distinctive segments prior to magnetization saturation. At the first stage, the

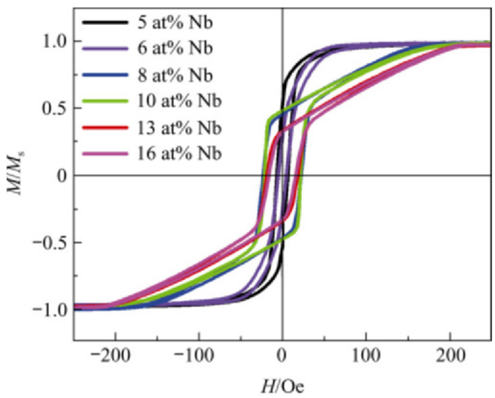


Fig. 2 In-plane hysteresis loops for room-temperature deposited FeCoNbB films with different Nb contents.

alloy is quickly magnetized at lower fields, while at the second stage the magnetization is linearly increased with magnetic field until the magnetically saturated state. The particular shape of hysteresis is called transcritical loop [18], which is typically seen in a film exhibiting perpendicular anisotropy [1,15,18]. Therefore, it can be considered that an out-of-plane anisotropy component is induced in the films with Nb content no less than 8 at%, which is in an amorphous state as shown in Fig. 1.

Fig. 3(a) presents variations of saturation magnetization ($M_s$) with Nb contents. It is seen that saturation magnetization exhibits an almost linear decrease with contents of Nb. The results can be explained by dilution of atomic magnetic moments due to the addition of non-magnetic elements [19-20]. Furthermore, perpendicular anisotropy constant ($K_\perp$) for the films with no less than 8 at% Nb has been calculated on the basis of equation $K_\perp = M_s H_k/2$. Here, the anisotropy field $H_k$ can be regarded as saturation field [12]. Fig. 3(b) shows dependence of $K_\perp$ on Nb contents. For the films exhibiting no transcritical loop, $K_\perp$ is defined as zero. It is found that $K_\perp$ decreases with increasing Nb contents for the films containing 8 at%-16 at% Nb, although the $H_k$ shows a slight increase as observed from Fig. 2. It can be therefore considered that the decrease of $K_\perp$ is mainly related to the linear decrease of $M_s$ with increasing Nb contents.

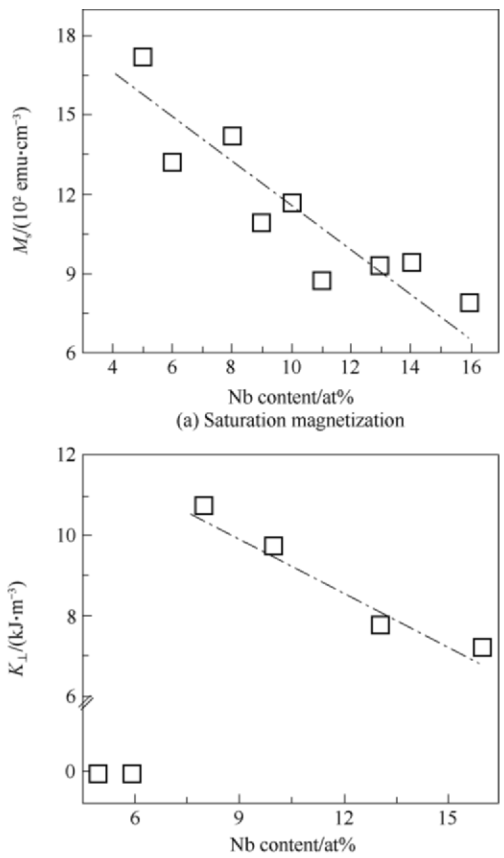


Fig. 3 Variations of saturation magnetization and perpendicular anisotropy constant with Nb contents.

### 3.3. *Origin of the out-of-plane anisotropy*

It is known that magnetic properties are determined by the competition among exchange, dipolar, and anisotropy energies. In films, when the perpendicular anisotropy energy is large enough to overcome the demagnetization energy, the magnetization vector will rotate from in-plane to out-of-plane. This phenomenon is the so-called spin reorientation transition (SRT) [21]. For an amorphous film, due to the absence of magnetocrystalline anisotropy, magnetic anisotropy is generally considered to be contributed by magnetoelastic coupling effect in thin films [11,15]. The magnetoelastic energy constant $K$ is expressed as $K=-3\sigma\lambda/2$ [15], where $\sigma$ and $\lambda$ represent stress and saturation magnetostriction constant, respectively, thus, orientation of easy axis depends on the sign of $K$.

It has been reported that FeCoNbB alloys exhibit positive magnetostriction constants in amorphous structure, and the magnetostriction constant is increased with the increase of Nb contents [20, 22-23]. From XRD analysis as shown above, it can be assumed that compressive stress is induced by lattice expansion in the alloys with Nb contents larger than 7 at%. On the other hand, it has been reported that the films deposited at lower argon pressure are likely to be in a compressive state [24]. Combining with the fact that out-of-plane component is observed only in the films deposited at a very low argon pressure of 0.8 Pa instead of 1.0-1.5 Pa, the out-of-plane anisotropy in FeCoNbB is very likely to arise from positive $\lambda$ and compressive stress. Furthermore, stresses induced in the films may be associated closely with Nb addition and/or preparation processes. It has been obtained that compressive stress is induced by lattice expansion in the alloys with Nb contents larger than 7 at%. In addition, as shown in Fig. 4, spin reorientation transition occurs only at lower argon pressure of 0.8 Pa instead of 1.0, 1.3 Pa. It

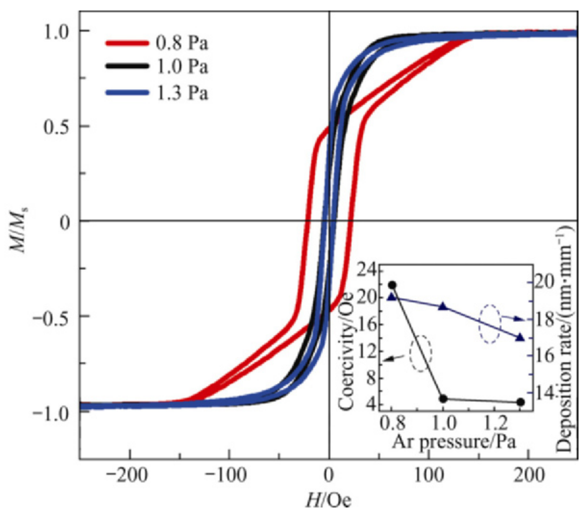


Fig. 4 Effect of Ar pressure on in-plane hysteresis loops for FeCoNbB films containing 10 at% Nb. (Inserted are variations of coercivity and deposition rate with Ar pressure.)

is also seen from the inset of Fig. 4 that the coercivity shows a quick increase once the spin reorientation transition from in-plane to out-of-plane occurs. As a consequence, magnetic softness is greatly deteriorated for the films.

The dependence of argon pressure on stress state in films is associated with variations of deposition rate. As shown in the inset of Fig. 4, deposition rate increases from 16.8 nm/min to 19 nm/min as the pressure decreases from 1.3 Pa to 0.8 Pa. Collision between sputtering particles is less frequent at lower pressure, resulting in an increase of free path for sputter ions. Therefore, more $Ar^+$ ions can reach the target at lower pressure. Furthermore, deposition pressure has a significant influence on stresses of films [24-25] due to variations of deposition rate, which gives rise to defects and induced compressive stress. The argument is consistent with results obtained by Dector [24].

As discussed above, it can be considered that the out-of-plane anisotropy in FeCoNbB is very likely to arise from positive $\lambda$ and compressive stress.

### 3.4. *Effective ways of eliminating the out-of-plane anisotropy*

In order to eliminate effect of the stress on the out-of-plane anisotropy induced during deposition, an amorphous film containing 13 at% Nb has been subjected to annealing in vacuum for 60 min at temperatures of 100, 150, 200 and 250 °C, respectively. No crystalline phase is detected by XRD for the films after annealing. As shown in Fig. 5, it is seen that the typical transcritical shape remains until 200 °C and vanishes for the film annealed at 250 °C.

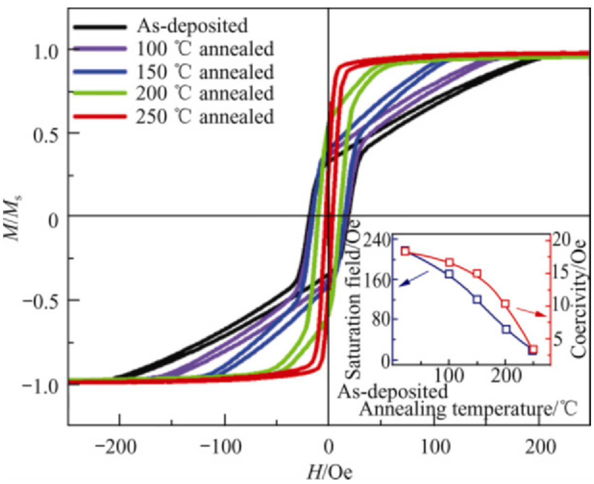


Fig. 5 Effect of annealing temperatures on hysteresis loops of FeCoNbB films containing 13 at% Nb. (Variations in saturation field and coercivity with different annealing temperatures are inserted.)

In addition, the out-of-plane component is reduced with increasing annealing temperatures. In particular, hysteresis for the 250 °C-annealed film exhibits a typical shape of magnetic softness, demonstrating disappearance of the out-of-plane anisotropy due to stress relaxation. As shown in the inset of Fig. 5, saturation field is reduced with increasing annealing temperatures. On the other hand, it is seen that effect of annealing temperature on coercivity is quite similar to that of saturation field. Based on these results, it can be concluded that the out-of-plane component induced in the films deposited at room temperature is attributed mainly to compressive stress.

Thickness dependence of the anisotropy has been further investigated. In-plane hysteresis loops of FeCoNbB with different thicknesses are displayed in Fig. 6. For the films with thickness of 420 nm, hys-

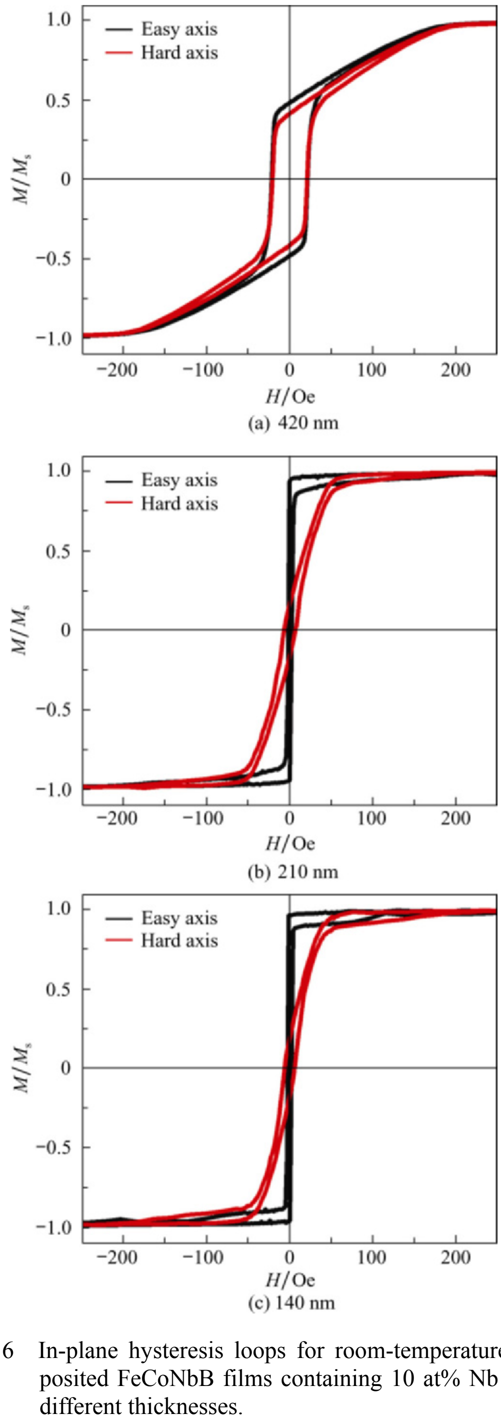


Fig. 6 In-plane hysteresis loops for room-temperature deposited FeCoNbB films containing 10 at% Nb with different thicknesses.

teresis loops along the easy and hard axes are superimposed, independent of direction of the applied in-plane field. The result is called rotatable anisotropy effect [26]. As film thickness decreases to less than 210 nm, square loop with in-plane uniaxial anisotropy is observed. In addition, coercivity along with in-plane saturation filed is decreased due to increase of demagnetization energy caused by reduction of thicknesses. Magnetic moment is in turn aligned with film surface.

As mentioned above, the magnetic properties are determined by the competition of different energies in films. In this work, competition is mainly between the demagnetization and perpendicular anisotropy energies. As the film thickness decreases, the two factors show different response: the former becomes larger while the latter decreases. As the film thickness decreases beyond a critical value, all magnetization vectors start to gradually rotate from out-of-plane to in-plane. Actually, film thickness has an effect on the film stress as well. As the thickness decreases, the compactness is decreased, which results in reduction of compressive stress [27].

Fig. 7 shows dependence of hysteresis loops on Si addition and field-deposition for the films. It is seen that deposition applied in a magnetic field has no influence on reduction of the out-of-plane anisotropy for the FeCoNbB films containing 8 at% Nb (see the dark and light blue lines). By contrast, together with Si addition, the film deposited in the external magnetic field exhibits a character of magnetic softness under the same applied magnetic field. Kohmoto, et al. [28] reported that addition of Si in FeCo-based amorphous alloys leads to a reduction of magnetostriction constant. According to $K=-3\sigma\lambda/2$ [15], the decrease of $\lambda$ leads to less negative values of $K$ and consequently magnetization vectors tend to align with in-plane of the films. The role of Si addition in reduction of out-of-plane anisotropy with field-deposition is considered to arise from magnetic ordering caused by presence of Si.

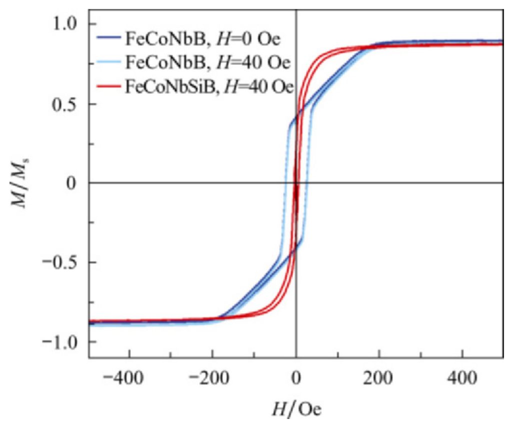


Fig. 7 In-plane hysteresis loops for films with different compositions and external magnetic field.

## 4. Conclusions

Crystalline and amorphous FeCoNbB films with various Nb contents have been prepared by DC-magnetron sputtering at room temperature. A structure transition occurs in the films with changing Nb contents. Furthermore, out-of-plane anisotropy component is observed in FeCoNbB with Nb contents larger than 8 at%. The unusual anisotropy is suggested to arise from stress-dependent magnetoelastic coupling effect. Thermal annealing, a reduction of film thickness and Si addition are found effective in eliminating the undesirable anisotropy and improving magnetic softness of the films.

## References

[1] Sharma P, Kimura H, Inoue A. Observation of unusual magnetic behavior: Spin reorientation transition in thick Co-Fe-Ta-B glassy films. Journal of Applied Physics 2006; 100(8): 083902-1-083902-10.

[2] Peng Y G, Ohkubo T, Laughlin D E, et al. The investigation of nanostructures of magnetic recording media by TEM. Scripta Materialia 2003; 48(7): 937-942.

[3] Takenaka K, Sugimoto T, Nishiyama N, et al. Structure, morphology and magnetic properties of Fe-B-Si-Nb glassy alloy thin film prepared by a pulsed laser deposition method. Materials Letters 2009; 63(21): 1895-1897.

[4] Li C Y, Duh J G. Magnetic properties of $Fe_{70-x}Co_{30-y}N_{x+y}$ thin films. Applied Surface Science 2005; 244(1-4): 477-480.

[5] Yu R H, Basu S, Zhang Y, et al. Pinning effect of the grain boundaries on magnetic domain wall in FeCo-based magnetic alloys. Journal of Applied Physics 1999; 85(9): 6655-6659.

[6] Hashimoto A, Hirata K, Matsuu T, et al. Crystal structure as an origin of high-anisotropy field of FeCoB films with Ru underlayer. IEEE Transactions on Magnetics 2008; 44(11): 3899-3901.

[7] Yu J, Chang C, Karns D, et al. Thermal annealing effect on FeCoB soft underlayer for perpendicular magnetic recording. Journal of Applied Physics 2002; 91(10): 8357-8359.

[8] Naohara T, The role of Nb in the nanocrystallization of amorphous Fe-Si-B-Nb alloys. Acta Metallurgica 1998; 46(2): 397-404.

[9] Sun H J, Man Q, Dong Y Q, et al. Effect of Nb addition on the glass-forming ability, mechanical and soft-magnetic properties in $(Co_{0.942}Fe_{0.058})_{72-x}Nb_xB_{22.4}Si_{5.6}$ bulk glassy alloys. Journal of Alloys and Compounds 2010; 504 (Sup.1): s31-s33.

[10] Jung H Y, Yi S. Enhanced glass forming ability and soft magnetic properties through an optimum Nb addition to a Fe-C-Si-B-P bulk metallic glass. Intermetallics 2010; 18(10): 1936-1940.

[11] Wang S X, Hong J, Magnetic and microstructural characterization of FeTaN high saturation materials for recording heads. IEEE Transactions on Magnetics 1999; 35(2): 782-787.

[12] Tiberto P, Celegato F, Coisson M, et al. Spin reorientation transition in amorphous FeBSi thin films submit-

ted to thermal treatments. IEEE Transactions on Magnetics 2008; 44(11): 3921-3924.

[13] Coisson M, Vinai F, Tiberto P, et al. Magnetic properties of FeSiB thin films displaying stripe domains. Journal of Magnetism and Magnetic Materials 2009; 321(7): 806-809.

[14] Sulitanu N. Structural origin of perpendicular magnetic anisotropy in Ni-W thin films. Journal of Magnetism and Magnetic Materials 2001; 231(1): 85-93.

[15] Chezan A R, Craus C B, Chechenin N G, et al. Influence of stresses and magnetostriction on the soft magnetic behavior of metallic films. Journal of Magnetism and Magnetic Materials 2006; 299(1): 219-224.

[16] Vázauez M, García, Prados C, et al. Magnetic anisotropies in single and multilayered thin films grown by bowed-substrate sputtering. IEEE Transactions on Magnetics 2000; 36(6): 3968-3974.

[17] Konno T J, Uehara M, Hirosawa S, et al. Effect of Nb addition on the crystallization behavior and magnetic properties of melt-spun Fe-rich Fe-Nd-B ribbons. Journal of Alloys and Compounds 1998; 268(1-2): 278-284.

[18] Alvarez-Prado L M, Alameda J M, Micromagnetism of nanowires with low out-of plane-anisotropy. Physica B 2004; 343(1-4): 241-246.

[19] Li S D, Liu M M, Yuan Z R, et al. Effect of Nb addition on the magnetic properties and magnetocaloric effect of CoMnSb alloy. Journal of Alloys and Compounds 2007; 427(1-2): 15-17.

[20] Chiriac H, Barariu F, Neagu M, et al. Effect of Mo and Nb additions on the magnetic properties of CoFeSiB amorphous wires. Journal of Non-Crystalline Solids 1999; 250-252(2): 762-765.

[21] Coisson M, Celegato F, Olivetti E, et al. Stripe domains and spin reorientation transition in $Fe_{78}B_{13}Si_9$ thin films produced by rf sputtering. Journal of Applied Physics 2008; 104(3): 033902.

[22] Betancourt I, Landa R. Magnetic properties of B-rich (Fe,Co)-Nb-B amorphous alloys. Journal of Alloys and Compounds 2009; 481(1-2):87-90.

[23] Palit M, Chelvane J A, Basumatary H, et al. The effect of Nb and Zr addition on the microstructural features and magnetic properties of $Tb_{0.3}Dy_{0.7}Fe_{1.95}$. Scripta Materialia 2009; 60(1): 56-59.

[24] Detor A J, Hodeg A M, Chason E, et al. Stress and microstructure evolution in thick sputtered films. Acta Materialia 2009; 57(7): 2055-2065.

[25] Han X L, Li W Z, Yuan K, et al. Analysis of low frequency SiN film on the effect of stress level and deposition parameters. Chinese Journal of Electron Devices 2010; 33(2): 139-141. [in Chinese]

[26] Saito N, Fujiwara H, Sugita Y. A new type of magnetic domain structure in negative magnetostriction Ni-Fe films. Japanese Journal of Applied Physics 1964; 19(7): 1116-1125.

[27] Shen B K, Wang Z H, Zhong Z Y, et al. Effeect of film thickness on the magnetic domain of $Co_{60}Fe_{20}B_{20}$, Journal of Chinese Electron Microscopy Society 2006; 25(3): 200-203.

[28] Kohmoto O, Ohya K. Amorphous FeCo-SiB alloys with zero magnetostriction. Journal of Applied Physics 1981; 52(2): 928-932.

## Biographies:

**YANG Youran** Born in 1985, she has received her B.S. degree from Beihang University in 2008 and now is a postgraduate student in Beihang University. Her main research interest lies in magnetic materials.
E-mail: yangyouran@yahoo.com.cn

**BI Xiaofang** Born in 1961, she has received her Ph.D. degree from Tokyo Institute of Technology in 1990 and now is a professor in Beihang University. Her main research interest lies in soft magnetic materials.
E-mail: bixf@buaa.edu.cn